PGDP

PADUCAH GASEOUS DIFFUSION PLANT

UNION CARBIDE

ASSESSMENT OF CONSOLIDATED UF₆
RELEASE STUDIES

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C. G. Jones

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September 7, 1983

COLLECTION Plant

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ASSESSMENT OF CONSOLIDATED UF6 RELEASE STUDIES

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PORTSMOUTH GASEOUS DIFFUSION PLANT

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Paducah Gaseous Diffusion Plant
Paducah, Kentucky
operated by
UNION CARBIDE CORPORATION
for the
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TABLE OF CONTENTS

| | · | Page No. |
|------|--------------------------------|----------|
| Ţ | EXECUTIVE SUMMARY | 1 |
| i • | RESPONSE TO DIRECTED QUESTIONS | 4 |
| II. | RESPONSE TO DIRECTED QUESTIONS | 9 |
| III. | SUMMARY | 11 |
| . V1 | REFERENCE | 11 |
| | BIBLIOGRAPHY | |
| VΙ | APPENDIX | 15 |

I. Executive Summary

The technical work addressed in this report had its origin in the technical support effort provided for the proposed packed-bed scrubber for the "Add-On" GDP at Portsmouth and a growing concern over the sites' ability to contain and predict the off-site impact of UF6 releases. the mid 1970s the technical managers agreed that scrubber studies would be conducted at Paducah and UF6 knockdown work at Portsmouth. result of the SAR effort and dispersion modeling studies conducted by Battelle Laboratories, Columbus, Ohio, the need for data on the basic chemistry and physics of the interaction of UF6 with atmospheric moisture was recognized. This assignment was directed to Oak Ridge in Prior to the consolidation effort of 1980, project action plans and objectives were developed with modest interaction between sites and without a directed set of objectives from management. Since that time, the Environmental, Health, and Safety (EH&S) Technical Program Management Team (TPMT) has insured against duplication of technical effort and provided project guidance on a limited scale. The authors recognize the wealth of information available in topically related technical reports. This report addresses the work in progress or initiated after The following project objectives the consolidation effort of 1980. (and their respective status) have evolved for the three sites.

A. PGDP

- 1. Objective: Characterize the performance of selected aqueous scrubbers in simultaneously removing UF6 and its hydrolysis products from gaseous streams.
- Status: A novel multichambered scrubber was found to remove both uranium and gaseous fluorides at greater than 99% efficiency under optimal conditions.

B. GAT

1. Objective: Determine the feasibility of various knockdown techniques for containing UF6 releases in feed, withdrawal, transfer and sampling areas. Characterization of the reaction between_UF6 and atmospheric moisture and evaluation of the variables of UF6 cloud formation and ultimate cloud fate have been consolidated to ORGDP.

 Status: Scoping type studies have identified electrostatic precipitation as the most effective means of particle knockdown.

C. ORGDP

- 1. Objective: Determine (1) the physical and chemical characteristics of UF5 hydrolysis products, (2) the mechanism of the hydrolysis reaction and the species and lifetime of the intermediates, (3) the morphology, change in morphology, and the rate of morphology change over a range of meteorological conditions.
- 2. Status: These objectives have been met over a number of release conditions where total UF6 released, humidity, temperature of UF6 at release, and release rate have been varied. The morphology of the UC2F2 formed has been characterized as a function of the conditions as has the change in morphology after release and the settling time of the cloud.

Major efforts have gone into the study of various types of UF6 releases, their plausibility, probability, and consequences. These UF6 release scenarios may be categorized as inside of structures or in the open. The release of UF6 vapor in an autoclave (estimated at about 80 15/min through a broken valve or pigtail) is considered Scrubbers tested under these a maximum credible occurrence. release conditions have proven capable of >99 percent removal efficiency for UF6 and its hydrolysis products and improving visibility in the area to a level where emergency efforts can commence. The desired concentration of uranium and HF in the scrubber effluent and the degree of operational flexibility required by operating groups would constitute the primary requirements for which scrubber design would be chosen for low assay areas. If the decision is made to employ scrubbers for this application, additional scrubber studies could be conducted to minimize the quantity of scrubber solution generated in the event of such a release. With studies in progress, the chosen scrubber design could also be challenged by higher inlet uranium concentrations and very low humidity condi-Any future scrubber work would require extensive renova-Severe equipment tion/upgrading of Paducah's test facility.

restrictions would be needed to adapt scrubbers for use in high assay areas. Although not evaluated to the extent that scrubbers have been, electrostatic filters have been demonstrated to be feasible at Portsmouth. Additional testing would be needed to optimize operating parameters for containment applications.

The most serious, credible releases defined by SAR work are those involving liquid-filled cylinders. A major scenario for the release of liquid UF6 inside a building (such as a ruptured cylinder in a feed facility) indicates that while the likelihood of occurrence is small, the potential impact to on-site and off-site personnel is great. Scrubbers are capable of significantly reducing the impact of this event.

For liquid releases outside a building, much can be learned from the release experienced at the French Comurhex Facility in July Upon breaking the valve (6 o'clock position) of a 7-ton cylinder at the threads (environmental conditions of 79°F, wind speed 20 mph), liquid UF6 spilled to the ground, with subsequent formation of a lingering dense cloud, for 15 minutes before a wooden plug could be inserted to stop the leak. The use of water spray was attempted for release control but seemed detrimental to the effort. A blanket of carbon dioxide was effective in knocking down the cloud, probably due to limiting access of atmospheric moisture to the UF6 and cooling effects, thus permitting access of Comparing these results to the worst case personnel to the area. scenario involving a 14-ton cylinder points to a significant concern over the control of an actual release and the time span The dense cloud limiting visibility and continued involved. release and reaction of UF6 point to the need for an acceptable means of cloud knockdown and reaction control for spilled material. This same concern over the availability of efficient methods to deal with outside releases of liquid UF6, was voiced as the first priority concern by Paducah Operations Division personnel when interviewed prior to preparation of this report. It is recommended that in addition to continued support of those release projects already budgeted and in progress at Portsmouth and Oak Ridge that funding and priority be provided to develop a set of "second generation" Techniques to handle outside releases of liquid UF6.

II. Response to Directed Questions

The following responses were prepared by the individual site members to the EH&S TPMT. These responses are directed toward the questions posed earlier by Donnelly.

- A. What were the original objectives?
- 8. Have the original objectives been met?
- C. Are the original objectives appropriate in today's atmosphere?
- D. Has the work significantly affected our ability to respond to a UFE release?
- E. Has the work significantly increased our understanding of the mechanisms of UF6 cloud behavior and knockdown?
- F. What additional work is desirable?

The site project objectives and their status are presented in greater detail in the Appendix.

G. PGDP

- The objective of the first phase of the scrubber effort at Paducah was to characterize the performance of selected aqueous scrubbers of the following types:
 - a. Wetted Fiber Pad (WFP)
 - Single-Stage Water Spray Chamber (SSW)
 - c. Novel Multichambered Scrubber (MCS)
 - d. Conventional Nozzleless Venturi Scrubber (NVS)

The characterization involved in simultaneously removing UF6, UO2F2, and HF from gaseous streams. A second phase of testing grew from the high removal efficiencies seen for the MCS system and the need to test a Conventional Nozzleless Venturi Scrubber (NVS). Second phase objectives included optimization of water consumption for the MCS, and evaluation of both systems (MCS and NVS) over a wide range of UF6 concentrations including insufficient moisture for complete UF6 hydrolysis.²

A second supportive area of study centered on the identification of a suitable electrolyte and sampling technique for UO₂F₂ particle size analysis by Coulter Counter methods.

These objectives have been met with the scrubber studies The MCS system was found to completed in September 1981. remove both uranium and gaseous fluorides at greater than 99 percent efficiency at optimum conditions. Attempts to reduce water consumption below a weight ratio of 1.8 kilograms per kilogram incoming air resulted in lowered efficiencies especially at very dry ambient conditions. The NVS system provided removal efficiencies (96.4 to 99.7 percent) slightly The NVC system proved to be more lower than that of the MCS. sensitive to changes in release conditions in that there is less flexibility for making operational adjustments. mance for the WFP was much lower for uranium and gaseous fluorides with removal efficiencies ranging from 69 to 98 percent. Application of the WFP system is thus marginal. Testing of the SSW system was terminated due to extremely poor performance as evidenced by penetration of the spray curtain and downstream deposition of UO₂F₂.

Suitable electrolyte systems for $U0_2F_2$ particle size analysis were identified as LiI/Isopropanol and LiBF $_2$ /Isopropanol. A major improvement in sample preparation was found to be collection of the sample on a filter medium followed by ultrasonic dispersion in the electrolyte.

- 3. The basic understanding of the capabilities of the various scrubber systems will stand as excellent core information. These studies provide confidence that off-the-shelf scrubbers remove particulate uranium at the same efficiency as they remove other particulate pollutants. Should employment of scrubber systems for GDP application be considered, a particular design can be chosen with confidence depending upon the required exhaust uranium concentration desired and the operational flexibility/sensitivity desired by the operating groups.
- 4. The work performed at Paducah, as defined by its objectives, was intended to enhance the capability of a GDP to respond to a UF6 release through the use of scrubber systems. Since scrubber employment is not currently considered, the value of the scrubber effort lies purely in its potential application.

- 5. The objectives of the scrubber studies conducted at Paducah did not address these issues.
- 6. Additional scrubber studies can be justified if a decision is reached to employ scrubber systems for GDP application. The final testing considerations are (a) possible recycle of scrubbing solutions to minimize the volume of liquid subsequently requiring treatment (b) extension of the studies to higher levels of uranium concentration. The potential value of solution recycle rests in the reduction of waste solution treatment and solid waste disposal costs. These potential savings would be considered in light of the projected infrequent use of the scrubbers and the known costs required to renovate/modify the scrubber test facility and conduct the required tests. As part of these studies the proposed scrubber design should be challenged by higher incoming uranium concentrations to simulate more adverse release conditions.

H. GAT

1. In response to recommendations resulting from containment problems and the increased emphasis in containment for employee and public safety, knockdown studies were initiated at The objective of this work was to determine the Portsmouth. feasibility of various knockdown techniques for containing releases in feed, withdrawal, transfer, and sampling areas. Associated with this effort were the tasks of characterizing the reaction between UF6 and atmospheric moisture and evaluating environmental variables of UF6 cloud formation and Due to delays in obtaining the environmental ultimate fate. chamber and changes in personnel, this work was not started until 1978 and completed in 1981. Current studies include activities in two areas. One deals with UF6 containment with the objective of developing techniques for mitigating, controlling, or eliminating existing or postulated credible accident hazards to plant personnel and/or public safety resulting from failure of plant components containing UF6. The other deals with UF6 release plume studies with the objective of providing assistance or plume tracking, analysis, and knockdown to the DOE/French information exchange team, primarily using the environmental chamber.

- 2. Within the scope of the original objective to determine the feasibility of various knockdown techniques, the objective has been met as electrostatic precipitation utilizing a charged stream of dry air were identified as the most effective. The associated task of characterizing the reaction between UFG and atmospheric moisture was consolidated to ORGDP and the task of evaluating environmental variables of UF6 cloud formation and ultimate fate was partially completed at Portsmouth with the remainder of this effort being conducted at ORGDP. Portsmouth, no association between settled UO₂F₂ cloud particle sizes and temperature or humidity effects in the range encountered in GDP operations were observed. The majority of particles analyzed were in the 0.5 μ to 3μ range. In spite of the high density of UO₂F₂ particulate material, UF₆ hydrolysis clouds were observed to rise followed by diffusion. Turbulence was observed to shorten settling time by enhancing agglomeration; however, this would not be expected in open air environments where current UF6 containment studies have dealt with evaluating commercial electrostatic air filters with reductions of 97 percent in airborne particulate concentrations being A rain simulation test has indicated little change Future activities in this in the airborne uranium content. project are to provide technical support and test environments for the development of fail-safe release alarm systems for cascade buildings and auxiliary facilities. UF6 release plume studies are being conducted to provide cloud settling rate data and establish means of seeing UF6 hydrolysis cloud growth, movement, and dissipation.
 - 3. The original objectives are appropriate since information about various knockdown techniques is essential for a feasibility study dealing with improved containment in areas with higher potential for failure of UF6 containing equipment. Objectives of current UF6 containment and release plume studies reflect near-term concerns and are revised as needed to respond to changes in anticipated needs.
 - 4. As defined by the objectives, the UF₆ containment work at Portsmouth was intended to evaluate the feasibility of various

knockdown techniques for containing releases. At this time, electrostatic concepts identified by this work are only candidates unique for future application in the GDPs.

- 5. Cloud behavior and the feasibility of various knockdown techniques, as defined by the objectives, are well understood for controlled environments. This information is applicable to small process areas where release clouds could be contained and knockdown techniques, as tested, utilized. Current UF6 release plume studies are providing information which should enhance our understanding of cloud behavior in open field situations.
- 6. Additional electrostatic filtering studies could be justified if a decision to install knockdown facilities such as electrostatic filters for GDP applications is made. Final testing would be needed to establish optimum operating conditions, nuclear safety parameters, unanium recovery techniques, and other operational considerations.

I. ORGDP

- 1. The original objective was to formulate a model that would accurately describe a UF6 release given the meteorological conditions, the amount of released UF6, and affected terrain. This model incorporated the various heats of reaction, association, and phase change. To supply empirical information to meet the needs of the model and to better understand UF6 releases, experimental work was performed at Portsmouth and ORGDP to determine (a) both the physical and chemical characteristics of hydrolysis products, (b) the mechanism of the hydrolysis reaction and species and lifetime of intermediates, (c) and the morphology, the change in morphology, and the rate of that change for a wide range of meteorological and total UF6 released conditions.
- 2. These objectives have been met over a number of release conditions where total UF6 released, humidity, temperature of UF6 at release, and release rate were varied. The morphology of the UO2F2 formed has been characterized as a function of the

conditions, as has the change in morphology after release and the settling time of the cloud. Intermediates between UF6 and U02F2 have not been observed under normal release conditions and, if they exist, their lifetimes are considered to be very short. The HF formed appears to weakly adduct to the U02F2, but is easily displaced by H20. The amount of hydration also is a variable yielding a large number of different U02F2 x H20 hydrates. A large effort was made to characterize the U02F2 x H20 formed, and to better understand the problems associated with its quantitative determination. The extent of hydration of HF and the onset of condensation as a function of HF and H20 concentrations were determined.

- 3. The original objectives are appropriate since the basic information about cloud makeup is essential to the understanding of cloud behavior and fallout.
- 4. Outside of considering several remote sensing techniques to follow a release, particularly after dispersion has rendered it invisible; no response has been considered other than the currently in place actions to ensure the safety of plant personnel.
- 5. Cloud behavior in a controlled environment is well understood.

 Much less is known about cloud behavior in the actual environment. Cloud behavior is dependent upon the characteristics of the aerosol particles composing the cloud; thus, a better understanding of cloud behavior in the actual environment exists when wind and dilution are considered.
- 6. More experimentation is needed to evaluate the early stages of aerosol particle formation and the effects of environment on that formation. In addition, conditions approaching ambient need to be tested to better determine how our current results minimize an ambient release.

III. Summary

The project objectives for the UF6 release work performed at Paducah, Portsmouth, and Oak Ridge are completed or currently in progress.

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These studies have been coordinated by the EM&S TPMT since 1980 to prevent duplication of technical efforts. It is desirable to continue the clume cloud studies in progress at Pontsmouth and the modeling studies under way at Oak Ridge. Further scrubber/electrostatic filtering studies are not warranted unless a decision favoring their employment is made. Additional scrubber studies would require extensive upgrading of Paducah's scrubber facility and an assessment of the resulting theatment/disposal requirements. Use of electrostatic filtering technology would require additional study to move beyond the feasibility stage and establish optimum operating conditions, uranium recovery techniques and other operational considerations. Scrubbers or electrostatic filtering would require evaluation of nuclear safety parameters.

Operations personnel were interviewed prior to preparation of this report to define their priority of needs concerning release technology studies. Technical support was requested in the following areas.

- A. Advanced technologies/procedures for handling liquid release of UF6.
- E. Training and equipment to make use of the plume dispersion model and topographical overlays in determining the off-site impact of UF6 releases.
- C. Updating the existing UF6 outleakage detection systems.
- D. Determination of means for egress from facilities under release conditions where visibility is impaired.

It is felt that in these areas can the greatest strides be taken to reduce the risk to on-site and off-site personnel as well as the environment.

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APPENDIX: STATUS OF SITE OBJECTIVES

This Appendix has been provided in order that a more extensive listing by site of the individual projects and their objectives and status can be examined. An attempt was not made to judge either the statistical design of the studies or the quality of the work performed. Rather the data and conclusions provided by the authors are taken at face value.

TABLE 1 PGDP

| Title/Document | Objectives | Status/Results |
|---|--|---|
| Evaluation of Aqueous Scrubbers for Removal of UF6 Hydrolysis Products. (KY/L-713, Part 1) | Characterize the performance of aqueous scrubbers: 1. Wetted fiber pad (WFP) 2. Single-Stage water spray chamber (SSW) 3. Novel multichambered device (MCS) in simultaneously removing UF6, UD2F2, and HF from gaseous streams. | Performance (removal efficiency) of the WFP varied from 69 to 98 percent and makes its application marginal. Inlet uranium concentrations for the trials ranged from 5000 mg U/m³ to 16,000 mg U/m³. The SSW was the poorest performer. Studies were terminated due to penetration of the spray curtain and downstream deposition of W2F2. The MCS performs comparable (>99 percent removal at optimum conditions) for UF6 systems as for other contaminants. |
| Advanced Aqueous Scrubber Studies (KY/L-1110, Part 9) | For the MCS: Test removal efficiencies over a wider range of inlet UF6 concentrations including insufficient moisture for complete UF6 hydrolysis. Optimize water consumption at atomizer chamber. | Over the relative humidity range of 15 to 75 percent, uranium renoval was not found to be a function of relative humidity. a. Renoval efficiencies remained above 99 percent for uranium and gaseous fluorides at optimum conditions. Concentrations for these tests range from 2660 mg U/m³ to 61,400 mg U/m³. |
| | | b. Reduction of water consumption belo |

a weight ratio of 1.8 kilograms per kilogram inlet air resulted in

lowered efficiencies.

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TABLE 1 PADUCAS (CONTINUED)

| Title/Document | Cb) ectives | Status/Pasults |
|---|--|--|
| ************************************** | c. Determine removal efficiency as a func- tion of particle size. | c. Insufficient data generated. Based on manufacturers projections & efficiencies seen, a mean particle size of 3µ was projected with 10% wt less than 1µ strongly suspected. |
| | d. Investigate possible efficiency losses at low inlet UF6 concentrations. | d. Over a wide range of ambient condi- tions and at low injet LF6 concentra- tions, remova! efficiences remained greater than 86.6 percent. |
| | Evaluate a conventional nozzieless venturi scrubber (NVS). | 2. The conventional nozzleless venturi scrubber provided removal efficiencies (96.4 to 99.7 percent) slightly lower than that of the MOS for uranium and gaseous fluorides. Selection for makin operating adjustments (i.e. fine tuning was not as great as for the MCS. These tests included uranium concentrations of 3,150 mg U/m³ to 44,700 mg U/m³. |
| UC ₂ F ₂ Particle Size Analysis by Coulter Counter Method (XY/L-725) | 1. Identification of a suitable electrolyte for U02F2 particle size analysis by Coulter Counter Model Tall. | 1. a. Suitable electrolytes were found to be L!!/isopropanol and LiBF4/ isopropanol. Both systems can be used with 50µ operature tube. |
| | 2. Determine feasibility and workability of particle size analysis of U072 in the lute of u range by the Coulter Counter Model Tall using identified electrolyte. | b. Exclusion of water from the electrolyte system is essential. c. Solubility of U02F2 in these electrolytes is low and only of concern for <1u particles. |
| | 3. Determine suitable sampling technique for collecting samples of W2F2 particles from an air stream such that particle sizes are not aftered and solubility problems are not encountered. | 2. 2. U0F2 particle size ranged from -0.8u to 40u. b. Predominance of particles in the 0.8u to 2.5u range. c. Reduction of instrument noise can |

be achieved by the placement of shielding around the sample beaker.

APPENDIX

TABLE 1 PADUCAH (CONTINUED)

| Title/Document | Objectives | | Status/Results |
|----------------|------------|----|--|
| | | | d. Additional study is needed to improve techniques for sealing sample beaker from humidity during analysis. |
| | | 3. | Samples should be taken on filter media and then dispersed ultrasonically in electrolyte. |

APPENDIX

TABLE 2 POSTSYOUTH

| Title/Document | | Colectives | | Status/Results |
|---|---------------------------------|--|---|--|
| UF6 Containment Studies | 1. | Characterize the reaction between UFG and almospheric | 1. | Effort consolidated to ORGOP |
| | | mois ture. | 2. | a. No association between UO ₂ F ₂ cloud particle size distribution and ten- |
| (GAT-T-3124, Part 6) (Mar. 1982) | ration and ultimate cloud fate. | | perature of humidity effects in range encountered in GDP operations. Majority of cloud particles in 0.3µ to 0.5µ range. Conclusions are for settled particles and many not apply | |
| | 3. | Evaluate use of water, | | to airborne particles. |
| | | steam, CaO ₂ , Freon, air jet, boric acid, and air on UF ₆ cloud knockdown and localization. | | b. Turbulence in chambers may shorten settling time by enhancing agglomenation. Opposite may occur in open field with cloud distribution. |
| | | • | | c. In spite of higher Sensity of U0272 particulate matter, UF6 release hydroloysis cloud immediately rises to ceiling of chamber followed by diffusion to fill chamber. |
| | | | 3. | Reproducible UF ₆ release cloud knockdown demonstrated (clear chamber in 5 minutes) with electrostatic-charged stream of dry air. Undisturbed cloud required 12 hours to 16 hours to settle |
| Positive UF6 Containment | 1. | Develop techniques for mitigating, controlling, or eliminating existing or postulated credible acci- dent hazards to plant per- sonnel and/or public safety | 1. | in progress - Feasibility of commercial electrostatic air filter demonstrated. Proposed use would be centralized loca- |
| (520 Subdivision Project Status File) | | | | tion with ducting to remove contaminate air from anticipated release areas. |
| (Thru May 1983) and (GAT-T-3124, Parts 1,4 5,6,7,8,10,£12) (FY-1982) | ι, | resulting from failure of plant components containing UF6. | | a. Material balance indicated 40 to 60 percent of release recovery with 80 to 97 percent accounted for. |

APPENDIX

TABLE 2 PORTS/OUTH (CONTINUED)

| Title/Document | Objectives | Status/Results |
|---|--|---|
| | • | b. Indicated 96.8 percent reductions in measurable particulate U0 ₂ F ₂ in air returned to chamber. Rain tests indicate little change in airborne Content. |
| UF ₆ Contairment Studies | Provide assistance on plume 1. tracking, analysis and | In progress |
| States | knockdown to DOE information exchange team primarily using the environmental chamber. | a. Differential sampling point, ver- tical profile sampler and timed dropout samples collected. No significant concentration dif- |
| UF ₆ Release Plume Studies | a. Determine cloud settling rates. | ference encountered through one foot differential sampler. Vertical profile sampler indicated |
| (520 Subdivision Project Status File) (Thru May 1983) | b. Establish means of seeing UF₆ hydrolysis cloud growth, movement, | gradual overall diminishing con- centration at all levels. Timed dropout sampler indicated 0-17 in/min settling rate. |
| · | and dissipation. | b. Ultraviolet, thermographic, and optical methods, tried in chamber were unsuccessful. Ultrasonic reflection and possible forward looking infrared scanner will be tried to track cloud. |

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TABLE 3 ORGEP

| Titie/Document | Cbjectives | | Status/Results |
|--|---|----|--|
| Determination of the Rate of 'F Hydration and the Effects of HF on Moisture Condensation (K/FS-155) Characterization of | Characterize solid products produced when UFg is released into air at specified relative humidities | | UF6 release experiments performed in small, static chambers at controlled atmospheric conditions; release chambers up to 8 cu. ft. in volume, UF6 release of 5 mg to 230 mg. RH of 2 to 100 percent. |
| the Solid, Alrborne Naterials Cheated by the interaction of UFG with Atmospheric Moisture in a Con- tained Volume (KP/S-144 | ·) <u>a</u> | 2. | Solid products were collected and characterized by: electron microscopy, x-ray diffraction, laser light scattering, and cascade impactor using mass measurements. |
| Characterization of the Solid Product(s) Formed When UF6 is Released into Ambient Air in a Contained Volume (K/ET-503) | 5. \$ | 8. | Various morphologies and several compounds were observed dependent upon conditions of release. The particle size and degree of agglomeration were dependent on the relative himidity of the air and the temperature of the UF6 at time of release. |

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